L. Hollberg

Time and Frequency Division National Institute of Standards and Technology Boulder, CO. 80303

Semiconductor diode lasers are emerging as important tools for the future of laser spectroscopy and precision optical measurements. The technology of diode lasers has advanced considerably in the past few years as has their availability. Because these lasers are practical, efficient and inexpensive they will open many research possibilities. Their low cost is particularly significant for laboratories with limited budgets. The impact of diode lasers on atomic and molecular spectroscopy will be profound, and is just beginning.

In this short summary of the characteristics of diode lasers and their application to spectroscopy we will focus attention on the room temperature semiconductor lasers operating in the near infrared and red regions of the spectrum. This means we will neglect completely the considerable amount of spectroscopic work that has been done with the cryogenically cooled lead-salt diode lasers that operate further in the infrared. The lasers we consider here are based on the mixed semiconductors of GaAs and InP and are produced primarily for commercial electronics applications including laser printers, compact disk players, and fiber-optics communications systems. Our goal is to take these commercial lasers and apply them to scientific and measurement applications. Some of the more general articles on the characteristics of diode lasers¹⁻⁴ and their application to spectroscopy⁵⁻⁷ are included in the references.

Semiconductor diode lasers are very <u>small</u>, electrically efficient, tunable sources of laser radiation that can provide reasonable cw power levels. They also have the potential for very high resolution spectroscopy. The diode lasers are semiconductor devices with dimensions of about 125x300x250 $\mu \rm m$ with an active laser region of about .3x3x250 $\mu \rm m$. The cleaved facets of the laser chip can serve as the mirrors for the laser's resonator because the index of refraction of the semiconductor is about 3.5 which gives a Fresnel reflectance of about 30%. Even with this small active region and mirror reflectance near 30% these lasers produce tens of milliwatts of cw optical power. The gain is obviously very high.

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The laser light is generated by the injection current which forces carriers through the active region. The fluorescent output power of the diodes increases gradually as the injection current is increased up to the threshold current, at which point the diode begins to lase and output power increases rapidly as a function of the injection current. A typical laser might have a threshold current of 50 mA (with a diode voltage drop of about 1.7 V) and a maximum output power per facet of 10 mW at 80 mA. Many of the new higher power lasers use high reflectance coatings on the laser's back facet and reduced reflectance coatings on the laser's output facet, so that all of the useful power comes out in one direction.

The performance characteristics that one can obtain from commercial diode lasers are diagrammed in fig. 1. Here we see the output powers that are available as a function of wavelength, with lasers available in four basic wavelength bands. The wavelengths are determined by the bandgap of the material; thus the shortest wavelength lasers (670 nm) are made from the semiconductor InGaAlP, those near 800 nm are made from AlGaAs, and the longer wavelength lasers come from various compositions of InGaAsP. The AlGaAs lasers generally have the best characteristics in terms of power and spectral purity, and they have naturally seen more applications in spectroscopy. We have used lasers from many regions of this chart but that is not to imply that it is necessarily easy to obtain lasers at any power and wavelength that one desires. Most of the lower power lasers are readily available from distributors but wavelength selection can be a problem. As a rule of thumb the price generally increases with power and wavelength.

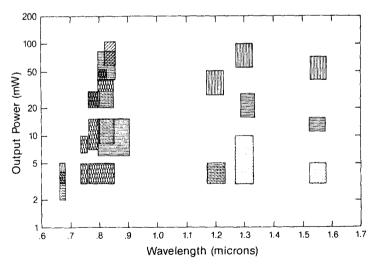


Fig. 1. Output powers for commercial semiconductor diode lasers plotted as a function of wavelength. The hatched boxes represent the distribution of lasers as advertised by various manufacturers.

A given laser's wavelength can be tuned easily with temperature and injection current. For example the tuning coefficients for AlGaAs lasers (for small temperature changes) are approximately +0.07 nm/K (or -30 $_{\rm GHz/K}$) and +0.007 nm/mA (or -3 GHz/mA) respectively. The tuning with the injection current is mainly due to heating of the semiconductor junction although there is a contribution due to changes in the carrier density. For larger temperature changes the laser's frequency will jump progressively from one longitudinal mode to the next. When one averages over these mode jumps and the linear tuning regions near each mode the net tuning rate with temperature is about 0.1 nm/K. But because of the mode jumps the spectral coverage is not complete. The pattern of mode jumps is usually irregular and often shows hysteresis, so the probability that a given laser will reach a specific wavelength within its tuning range is only about 50%. With a Peltier element for temperature control. a typical laser will cover (incompletely) a wavelength range of about 8 nm. For spectroscopic applications it is obviously wise to have several diode lasers available. In practice the problem is not very serious hecause diode lasers are inexpensive (some as low as US \$20) and in addition we can use optical feedback techniques to achieve almost complete spectral coverage within a wavelength range of about 20 nm.

Before applying diode lasers to spectroscopy we will find it useful to have some understanding of their noise characteristics. We need to consider both their amplitude and frequency noise. The amplitude noise of unmodified commercial diode lasers is broadband and extends from the audio frequency range to just above the laser's relaxation-oscillation frequency at a few gigahertz. The magnitude of the noise over this frequency range is typically 20 dB above the fundamental shot noise limit, and peaks to higher noise levels at low frequencies (below 100 kHz) and at the relaxation oscillation frequency (about 3 GHz). To scale these numbers in terms of fractional amplitude fluctuations we recall that the shot noise corresponds to the purely statistical fluctuations of randomly distributed photons, and thus varies as the square root of the number of photons. For example, the fractional power fluctuation due to shot noise for a 5 mW laser beam at 800 nm is about 10^{-8} . Thus we can expect a typical diode laser to have fractional power fluctuations of 10⁻⁶, except at the lowest frequencies where the noise is higher. Generally the noise is that it decreases with an increase in injection current or a decrease in junction temperature. In fact, the amplitude noise on many diode lasers is low when compared to that of other spectroscopic laser sources such as dye lasers. This advantage of diode lasers allows high sensitivity absorption measurements to be made relatively easily.

The characteristics of the amplitude and frequency noise depend profoundly on any optical feedback that finds its way back into the laser. This is a disadvantage of diode lasers for some applications and care must be taken to avoid feedback. Feedback can be avoided by careful attention to optical layout, or when necessary by using some form of optical isolator (attenuator, quarter-wave plate and polarizer, or Faraday isolator).

The spectral properties of the frequency noise on diode lasers depends strongly on the type of laser. In fact, some commercial lasers do not even run on a single longitudinal mode, which makes them hard to use for spectroscopy. The longitudinal mode spacing for these very tiny lasers is about 0.3 nm (\approx 140 GHz) and the bandwidth of the gain is about

40 nm. As the technology advances more and more lasers that operate on a single longitudinal mode are available. Although there are optical methods that can be used to force multimode lasers to operate on a single mode, we will concentrate on the single mode lasers. The spectral linewidth of most commercial diode lasers varies from about 20 to 300 MHz. These linewidths provide a resolution that is adequate for some spectroscopic applications but not for others. The spectral character of the frequency noise that generates these broad linewidths is similar (and related) to the amplitude noise. That is, the spectral density of frequency fluctuations is broadband and extends out to tens of megahertz before it drops off significantly. There is then a broad flat region of frequency noise out to higher frequencies and again a peak in the noise at the relaxation oscillation frequency (at a few gigahertz). The frequency noise below 20 MHz is the most significant because it contributes the most to the linewidth.

One of the early surprises that people found when trying to use diode lasers for atomic spectroscopy was that the signal-to-noise ratio in some cases was not nearly as good as expected.8 It turns out that the frequency noise on diode lasers can be converted to amplitude noise by atomic resonances. This then degrades the signal-to-noise ratio in absorption and fluorescence measurements. In using diode lasers for optical pumping in cesium atomic clocks, people discovered that the noise on the strong cycling transition (F = 4 to 5) was about 100 times worse than expected. The physics of this FM-to-AM noise conversion by the atomic resonance has been explained, at least in part, by the work of Zoller and collaborators. 9 In fig. 2 we show some experimental manifestation of this noise in fluorescence and absorption measurements. In fig. 2a we see the output from a very simple experiment; the diode laser's output was sent through a cesium cell and then onto a fast photodetector. As the laser was scanned across the Doppler broadened transition, the high frequency noise at the photodetector increased dramatically. Figure 2b shows the Doppler free fluorescence spectrum of the cesium, F = 4 to 5, transition taken in an atomic beam. Here again we see a large increase in the noise from the atomic signal, whereas the Fabry-Perot transmission fringe that was monitored simultaneously shows mainly frequency modulation and not the excess amplitude noise. Fortunately, this type of noise is insignificant on many transitions and can be eliminated when it is significant by spectrally narrowing the lasers as described below.

Two main approaches have been taken to reduce the linewidth of semiconductor diode lasers: one is fast electronic feedback and the other is some form of controlled optical feedback. The electronic feedback approach uses fairly traditional laser frequency control to feed back to the laser's injection current in order to stabilize the lasers frequency to a Fabry-Perot resonance. With this method it is very easy to precisely control the laser's center frequency, but it is not easy to reduce the diode laser's linewidth. The trick here is that the frequency noise extends to high Fourier frequencies so that very fast electronics are required in the servo-loop. A few groups have had some success in narrowing diode laser's linewidths with fast electronics. 10-12

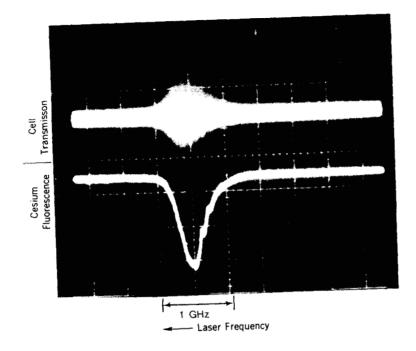
The other main approach to diode laser frequency control is to use some form of controlled optical feedback in order to narrow the laser's linewidth. The linewidth of a diode laser can be reduced simply by reflecting some of the laser's output back to the laser. This effectively extends the laser's resonator and creates a coupled cavity system with the original diode laser cavity coupled to an external reflector. This extended cavity system has a higher Q than the original diode laser cavity and thus produces a narrower linewidth. It is often

advantageous to have some frequency selectivity in the optical feedback to force the laser to a specific single longitudinal mode. A wide variety of optical feedback systems has been developed to control the center frequency and narrow the linewidth of diode lasers. For example, the feedback can come from mirrors, fibers, gratings, etalons or combinations of these. With many of these systems it is necessary to have an antireflection (or reduced reflection) coating on the diode laser's output facet. This reduces the competition between the normal diode laser modes and the extended-cavity laser modes. We will look with a little more detail at two optical feedback systems that have been very useful for spectroscopy.

One of the ideas that has existed for some time is to use a grating to extend the diode laser's cavity and thus provide an extended cavity with frequency selective feedback. 13-19 This system works very well if the laser has some antireflection coating on its output facet. Such grating systems can be used to tune the laser's frequency in steps over a wavelength range of approximately 20 nm. Fortunately (and fairly recently) many of the commercial high-power lasers already have antireflection coatings and can be used directly with gratings without modification. The grating then allows one to dependably select a specific longitudinal mode of the original diode laser chip. Small changes in the diode's temperature can be used for gross tuning of these modes. With stable mechanical construction these grating extended cavities have linewidths of 0.1 to 1 MHz depending on the actual design. With the grating mounted on a PZT translator the laser's frequency can be scanned continuously over a range of about 1 GHz. If the diode laser's injection current is scanned synchronously with the PZT the continuous scanning range can be tens of GHz.

Another frequency stabilization method that we have used successfully is an optical-feedback lock²⁰⁻²² that is based on resonant optical feedback from a high-Q optical resonator. A typical system of this kind uses an unmodified commercial diode laser and weak optical feedback from a confocal Fabry-Perot cavity. In certain geometries the laser sees feedback from the Fabry-Perot only when the laser's frequency matches the resonance frequency of the Fabry-Perot. In this coupled cavity system the laser's oscillation frequency automatically locks to the cavity resonances and is thereby stabilized. These optical self-locking systems have achieved diode laser linewidths of a few kilohertz.

One of the very useful properties of diode lasers is that they can be modulated very efficiently and very rapidly via the laser's injection current. When the injection current is modulated, the laser's power and frequency are both modulated. In practice, for spectroscopic applications, the variation in power is small relative to the variation in frequency and can often be ignored. The modulation response of the diodes extends from DC to a few GHz and allows a number of applications that are much more difficult with other types of lasers. Some of these applications include rapid frequency scans (frequency chirps) which can be used for diode laser cooling of atoms, 23 rapid frequency jumps which can be useful for transient spectroscopy and optical pumping, and high frequency modulation which can be used for optical heterodyne spectroscopy. Unfortunately, the modulation capabilities are altered ^{and} usually degraded by the optical stabilization methods that are useful for narrowing the laser's linewidth. This is not surprising because in one case we are asking the laser's frequency to be very stable and in the other we are asking it to change easily and rapidly.



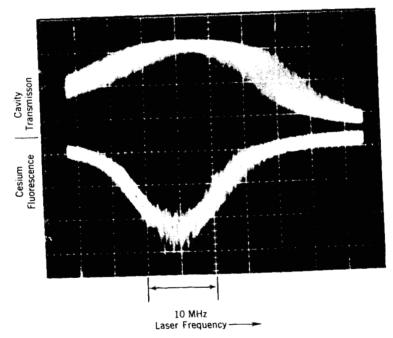


Fig. 2. Noise observed in cesium spectroscopy. In 2a the upper trace shows excess high frequency noise on a diode laser beam that has passed through an absorption cell. The lower trace in 2a is the fluorescence signal detected simultaneously. In 2b the lower trace shows excess noise in atomic beam fluorescence from the F=4 to 5 transition. Here the upper trace is a Fabry-Perot transmission fringe that was taken simultaneously.

To be realistic about applying diode lasers to spectroscopy and precision measurements we must also recognize their limitations. One of these is that some of the special diode lasers we would like to use are very difficult to obtain. In addition, some of the lasers that we can get do not tune to the wavelength we need, and their tuning is irregular with many mode jumps. For high resolution spectroscopy the diode laser's broad linewidths (tens of megahertz) are a problem. The diode lasers are also particularly sensitive to optical feedback. For some applications it would be useful to have more power than is readily available from common diode lasers. This brief list of complaints contains most of the limitations that we face when trying to use diode lasers, but the problems are not very serious. Relatively simple techniques have been developed to deal with the tuning, linewidth, and optical feedback problems.

On the other hand, the advantages of semiconductor diode lasers far outweigh their limitations. For the most part the lasers are inexpensive, electrically efficient, readily available, and extremely easy to use. Typical powers of tens of milliwatts are more than enough for even nonlinear spectroscopy. The diode lasers are easily swept and modulated which allows unique applications. With a little bit of extra work the laser's frequency can be stabilized and one can achieve resolution capabilities of a few kilohertz. The available diode lasers are produced for commercial electronics, and we are just in the early phases of learning how to use them for scientific applications. The development and application of diode lasers for spectroscopy is somewhat reminiscent of the early days when the transistor began to take over some of the roles of the vacuum tube. The technology is changing rapidly and the future certainly promises higher powers, broader spectral coverage and better spectral purity.

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